Comparison of HALOE and MLS HCl with Calculated Time Series from Several Photochemical Models

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E.-S. Yang [4], L. Froidevaux [1], T. Canty [1], J. W. Waters [1],
A. Douglass [5], E. Fleming [5], C. Jackman [5], S. Montzka [6], D. Waugh [7]

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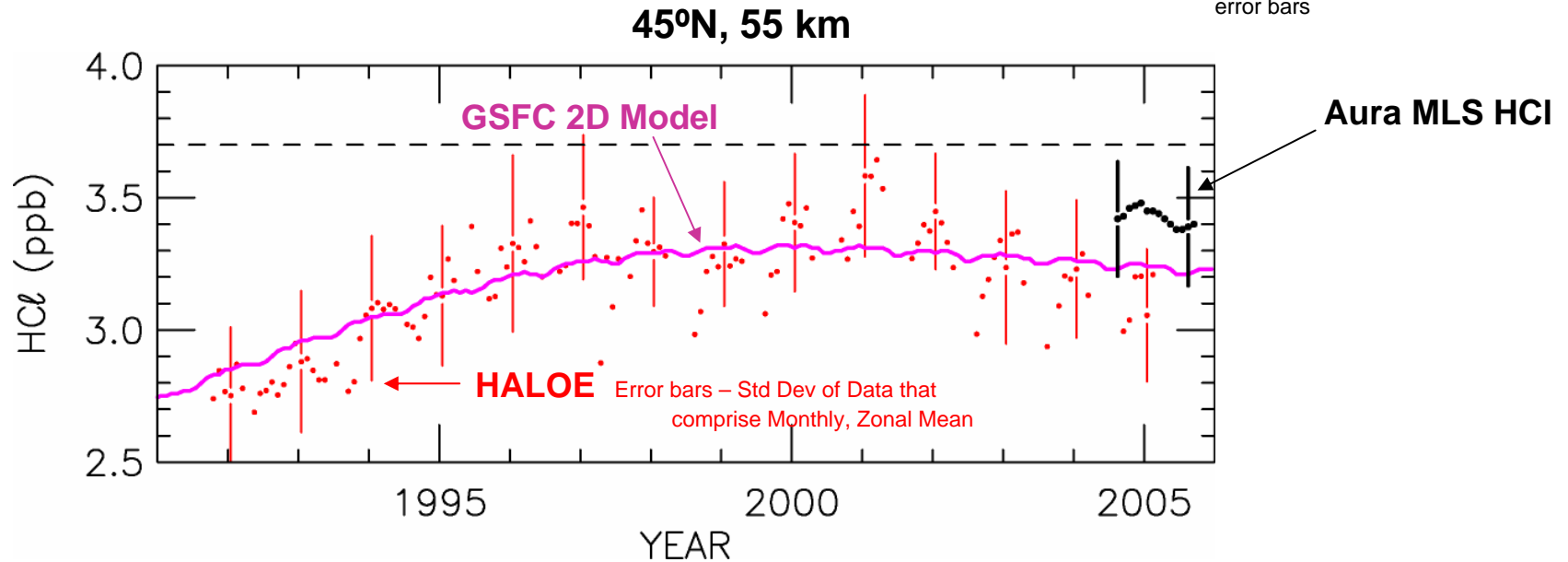[5] NASA Goddard Space Flight Center, Greenbelt, MD

[5] NOAA CMDL, Boulder, CO

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Introduction

45⁰N, 55 km

- Zonal, monthly mean HALOE HCl agrees fairly well with this 2D model calculation

- Model run: GSFC 2D model with climatological transport
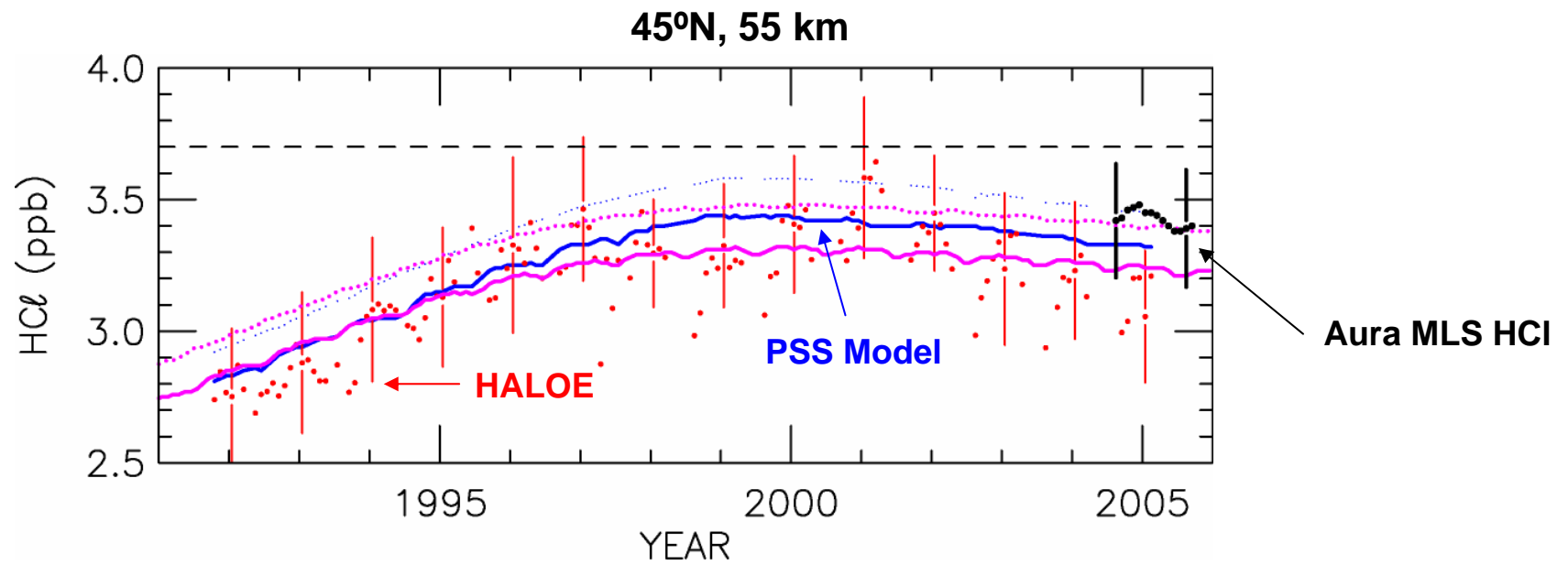 Jackman et al., JGR, 2005

- Focus on:

  Factors that govern this comparison

  - **age of air (<u>mean value</u> & <u>shape of distribution</u>)**

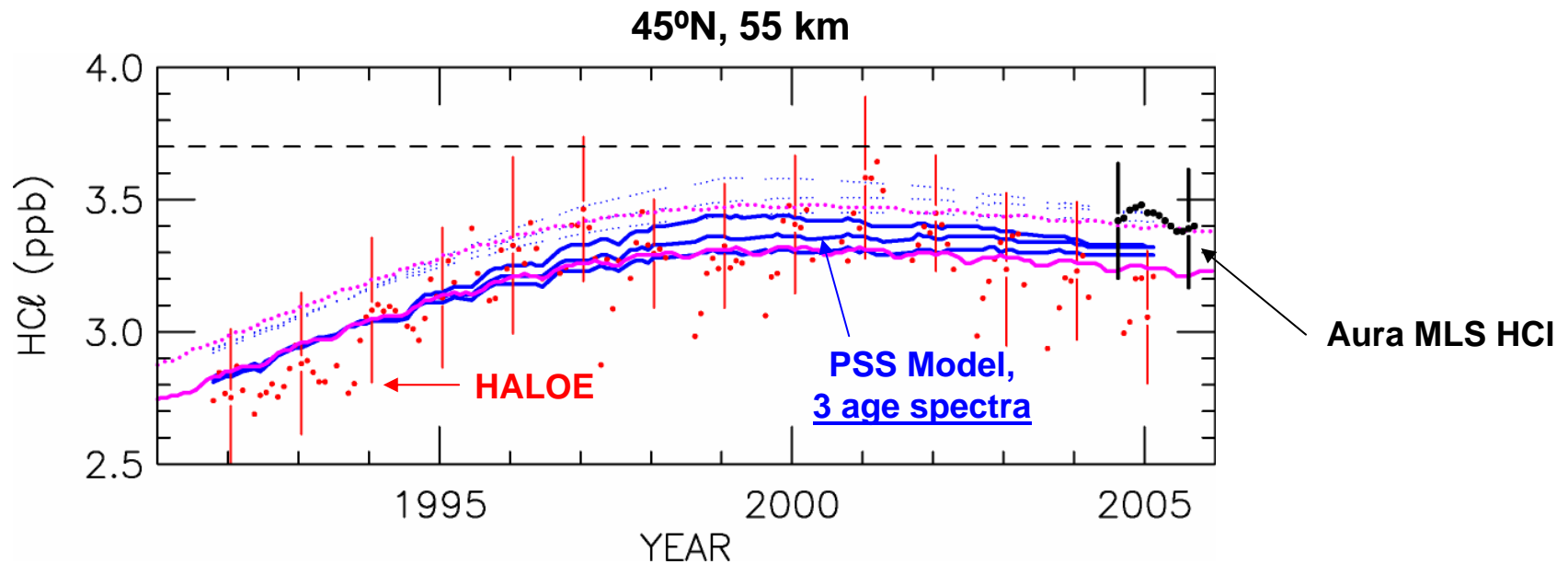 - **contribution from very short lived (VSL) organics to Cl$_y$**

Cl$_y$ appears as DOTTED LINES

45ºN, 55 km
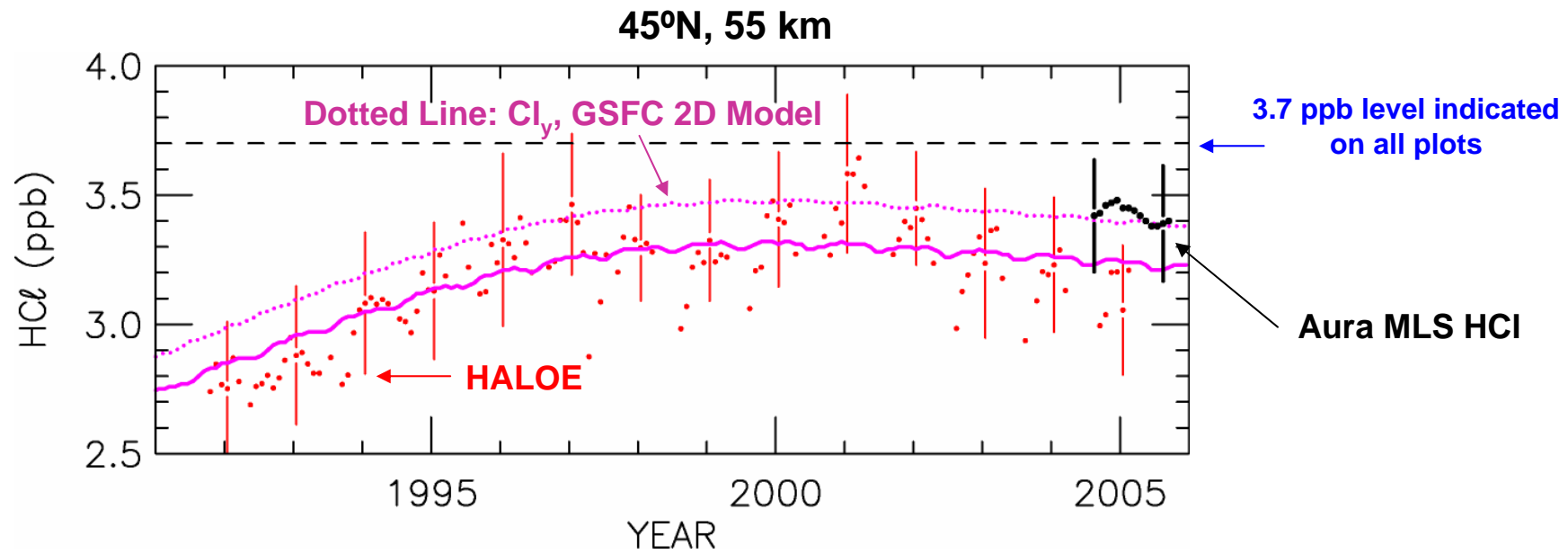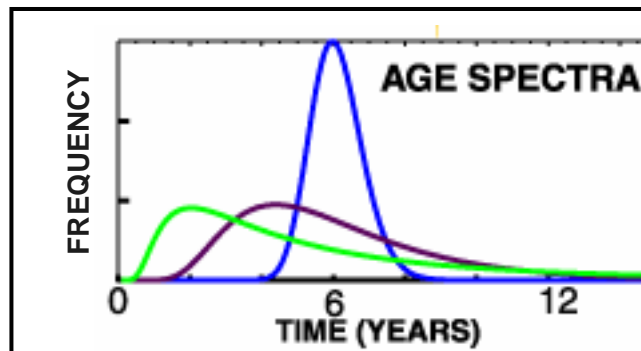
PSS Model Results appear as BLUE LINES

45ºN, 55 km

Three PSS Lines representing air with _Same Mean Age_, different "Age Spectra"

45⁰N, 55 km

HALOE

**PSS Model,
3 age spectra**

Aura MLS HCl

Three age of air spectra included for all PSS simulations:
- Mean age of 6 years
- Widths of 0.01, 2.0, and 3.0 yrs
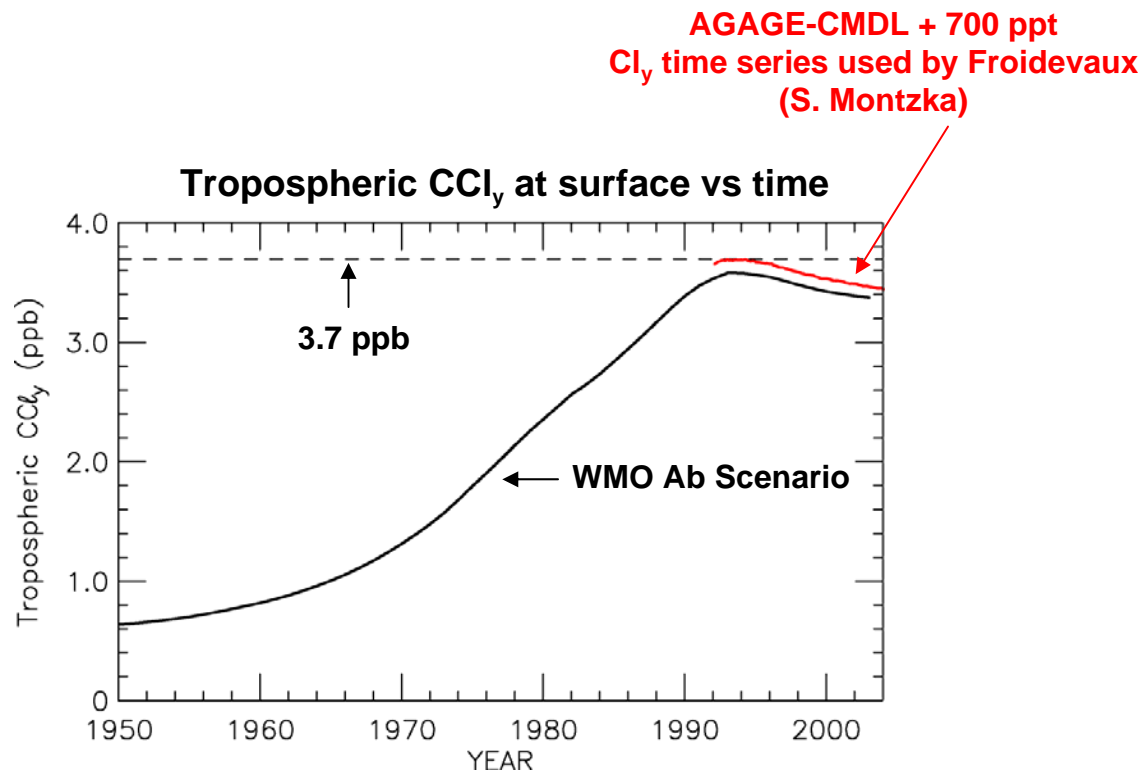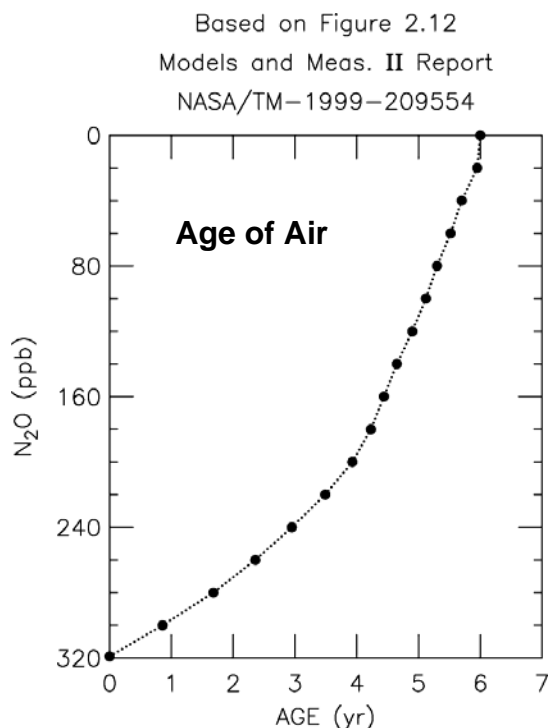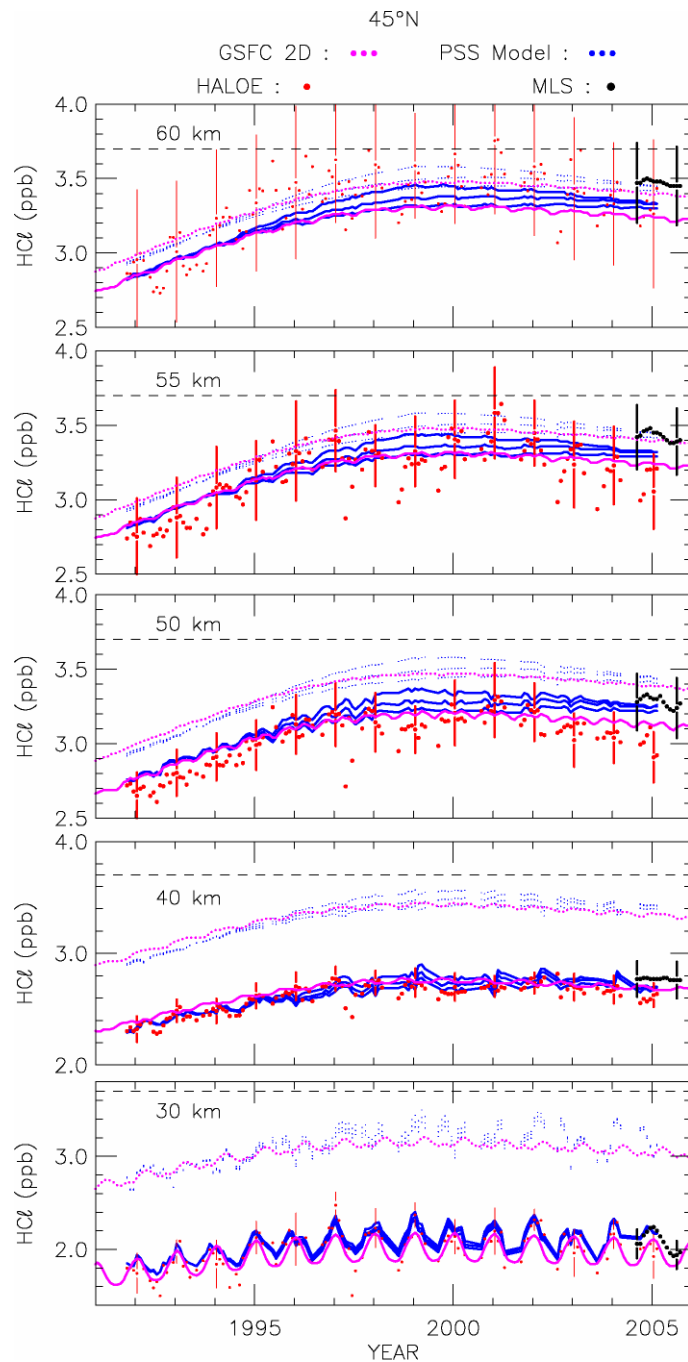

AGE SPECTRA

Waugh et al., GRL, 2001 :
- All distributions have mean age of 6 yrs
- Mean ages of 0.5, 2, and 4 years

PSS (Photochemical Steady State) Model

- HALOE CH_4 used to calculate N_2O (Michelsen *et al.*, *GRL*, 1998)
- Cl_y and NO_y estimated from N_2O using standard correlations
- O_3, Surface Area, H_2O, etc from HALOE and SAGE II
- dN_2O/dt, dCH_4/dt, Age of Air, and CCl_y vs time considered

AGAGE-CMDL + 700 ppt Cl_y time series used by Froidevaux (S. Montzka)

Based on Figure 2.12
Models and Meas. II Report
NASA/TM−1999−209554

Tropospheric CCl_y at surface vs time

Age of Air

3.7 ppb

WMO Ab Scenario

Same modeling approach used to analyze aircraft & balloon data (Salawitch et al., Osterman et al., Sen et al., Jucks et al., etc)
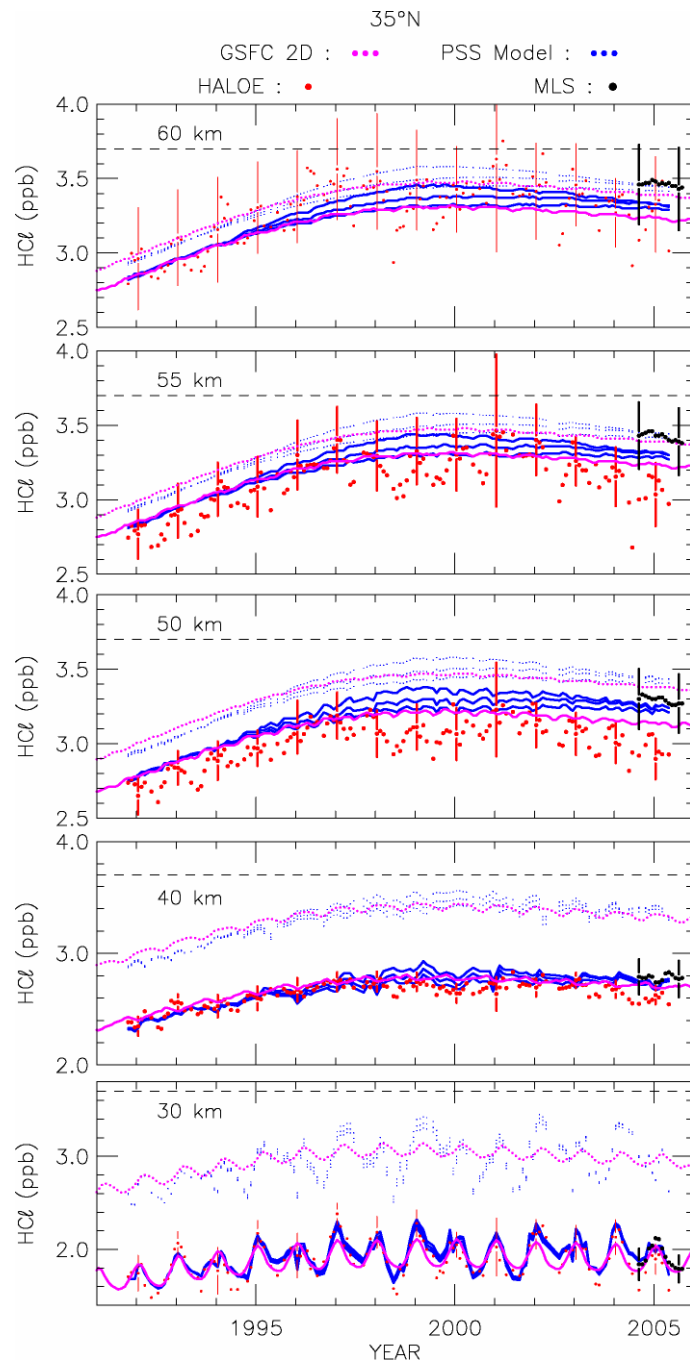
45°N

- GSFC HCl indicative of wide age spectrum for all altitudes
- Age spectrum more important ~1997 to 2002
- Models using WMO Cl_y underestimate MLS HCl above 40 km
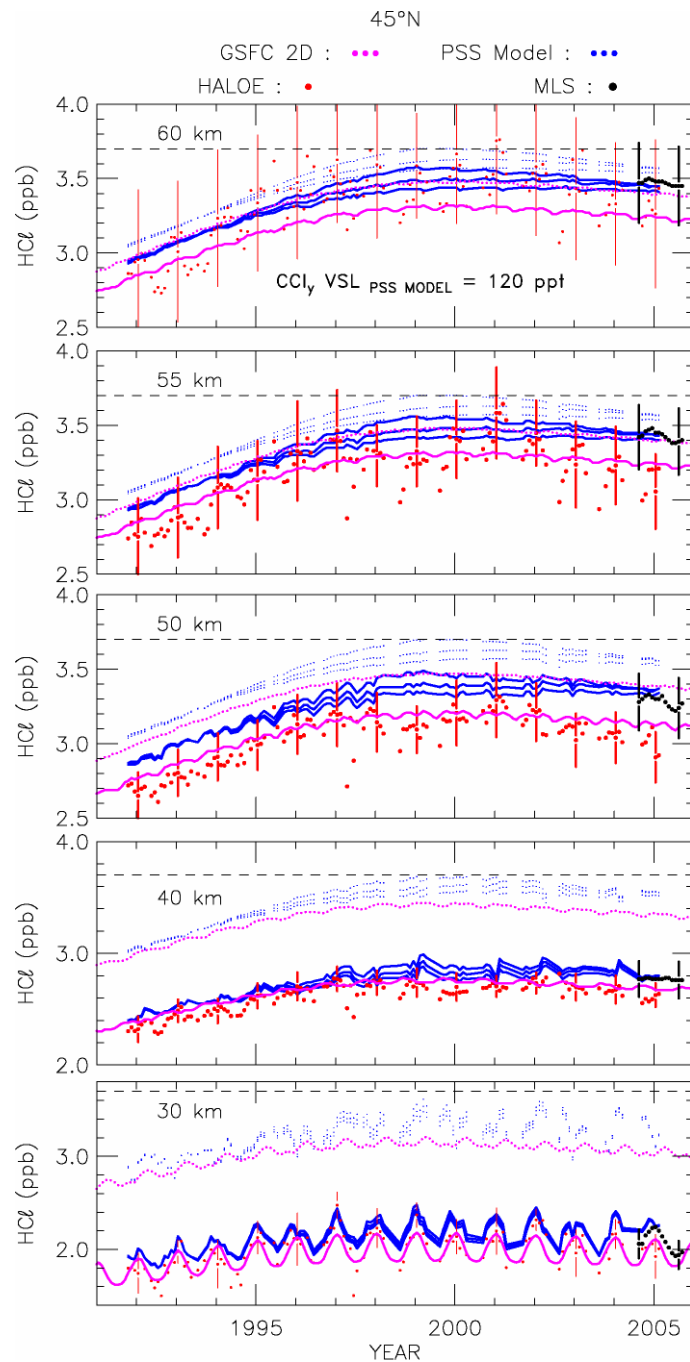
If VSL compounds contribute 120 ppt to Cl_y PSS model:

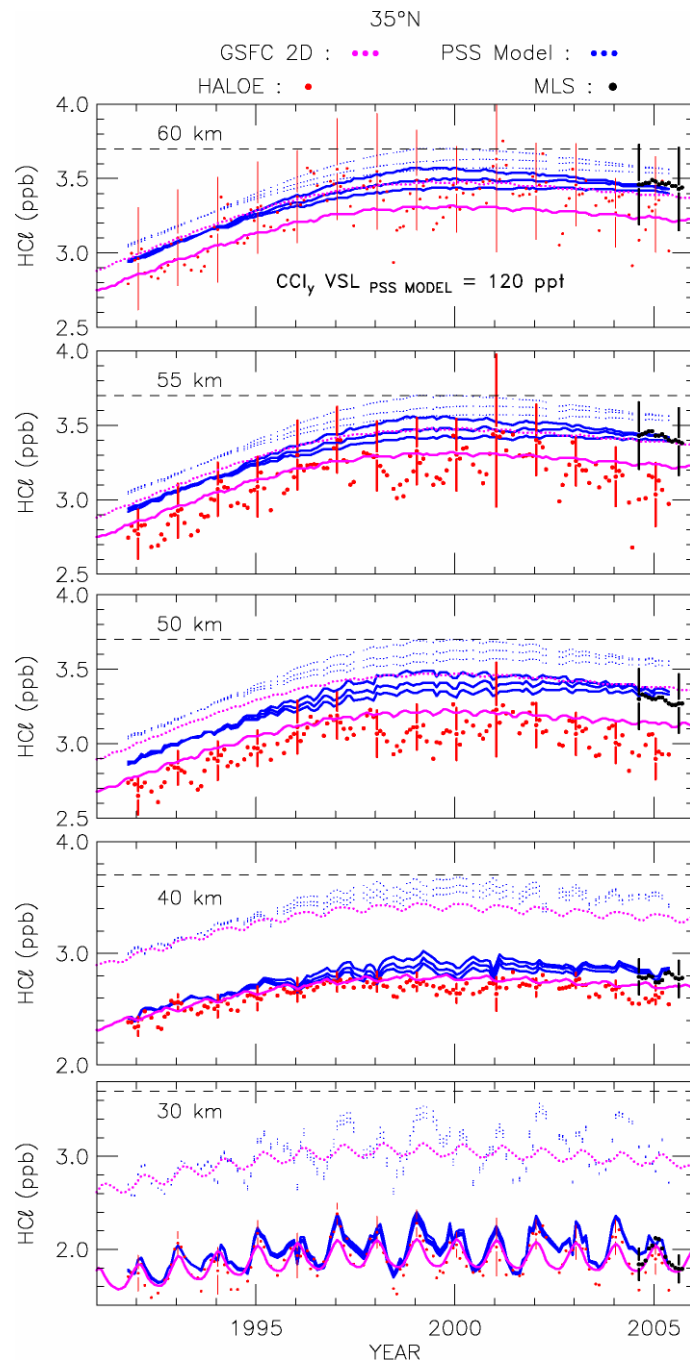– agrees well with MLS HCl all altitudes (mid 2004 to present)

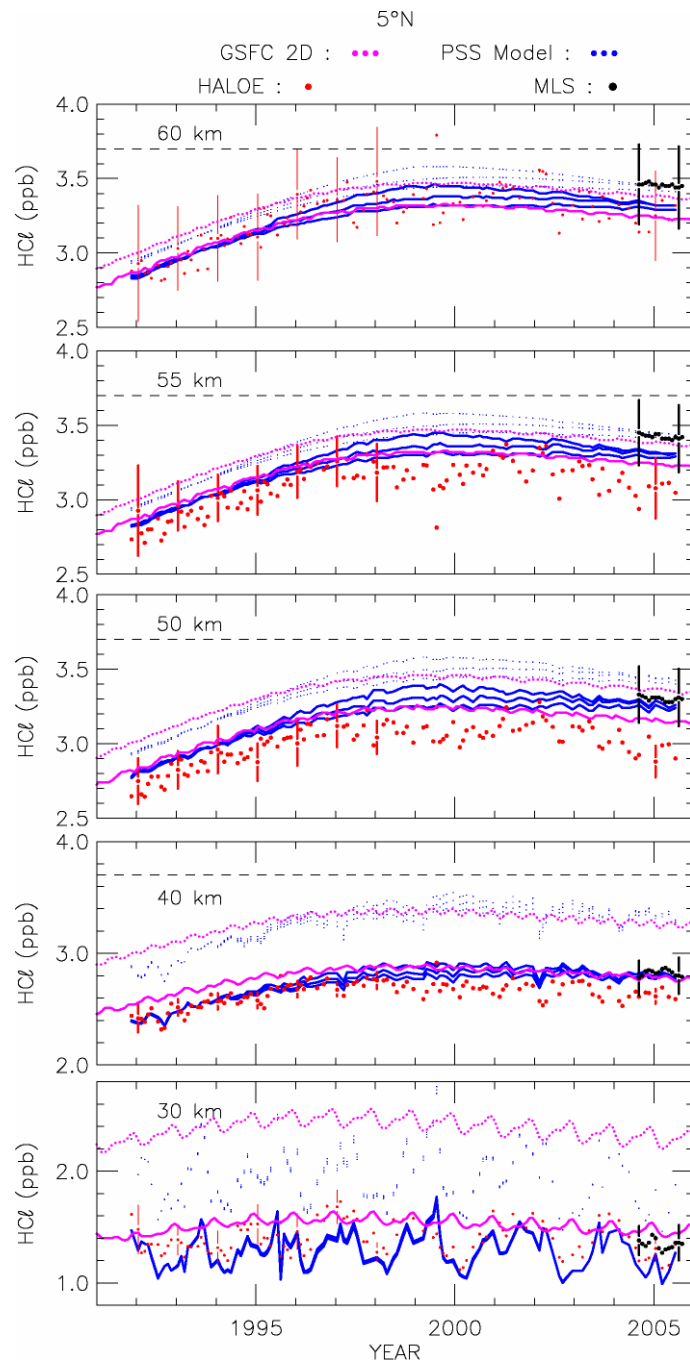– overestimates HALOE HCl all altitudes, most times
(1992 to present)

35°N

35ºN

– **GSFC model: WMO Cl$_y$**

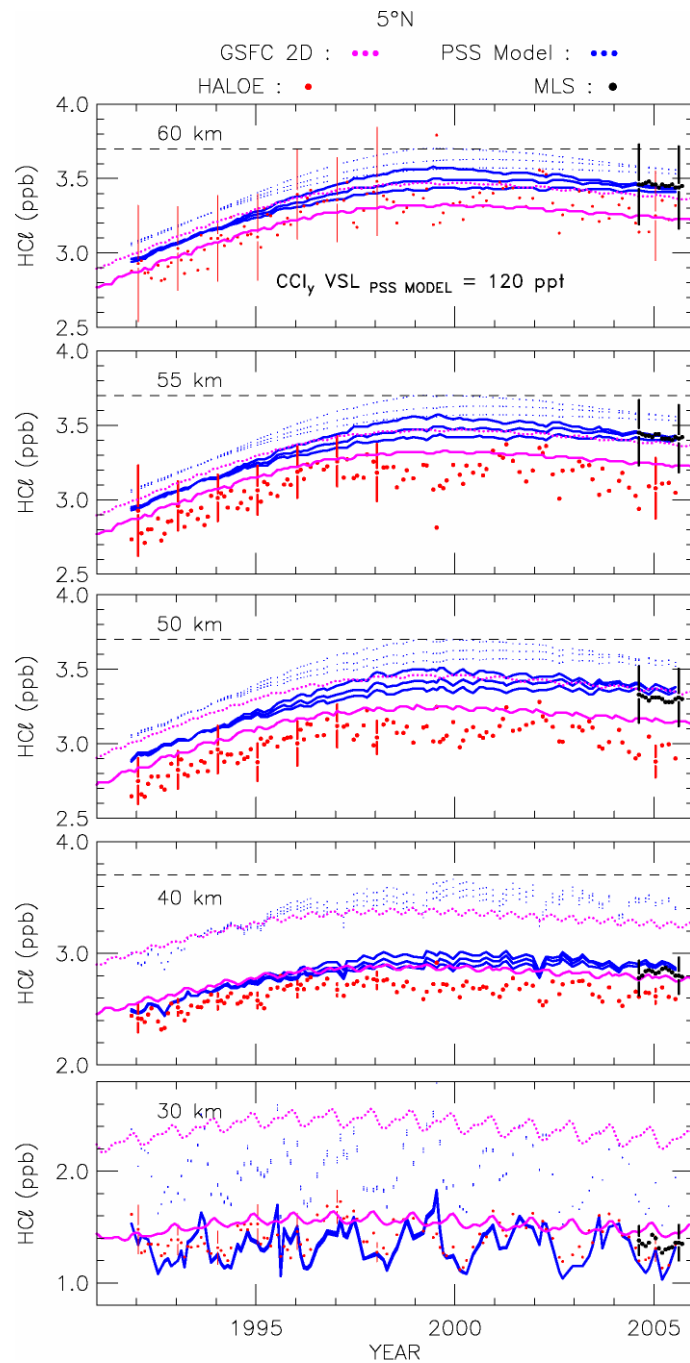– **PSS model: WMO Cl$_y$**

35°N

- GSFC model: WMO Cl_y
- PSS model: WMO Cl_y + 120 ppt

5°N

– **GSFC model: WMO Cl$_y$**

– **PSS model: WMO Cl$_y$**

5°N

GSFC 2D : ⋯ PSS Model : ⋯
HALOE : • MLS : •

60 km

$CCl_y VSL_{PSS\ MODEL} = 120\ ppt$

55 km

50 km

40 km

30 km

1995   2000   2005

YEAR

5ºN

– **GSFC model: WMO Cl_y**

– **PSS model: WMO Cl_y + 120 ppt**

5ºS

– GSFC model: WMO Cl_y

– PSS model: WMO Cl_y

5ºS
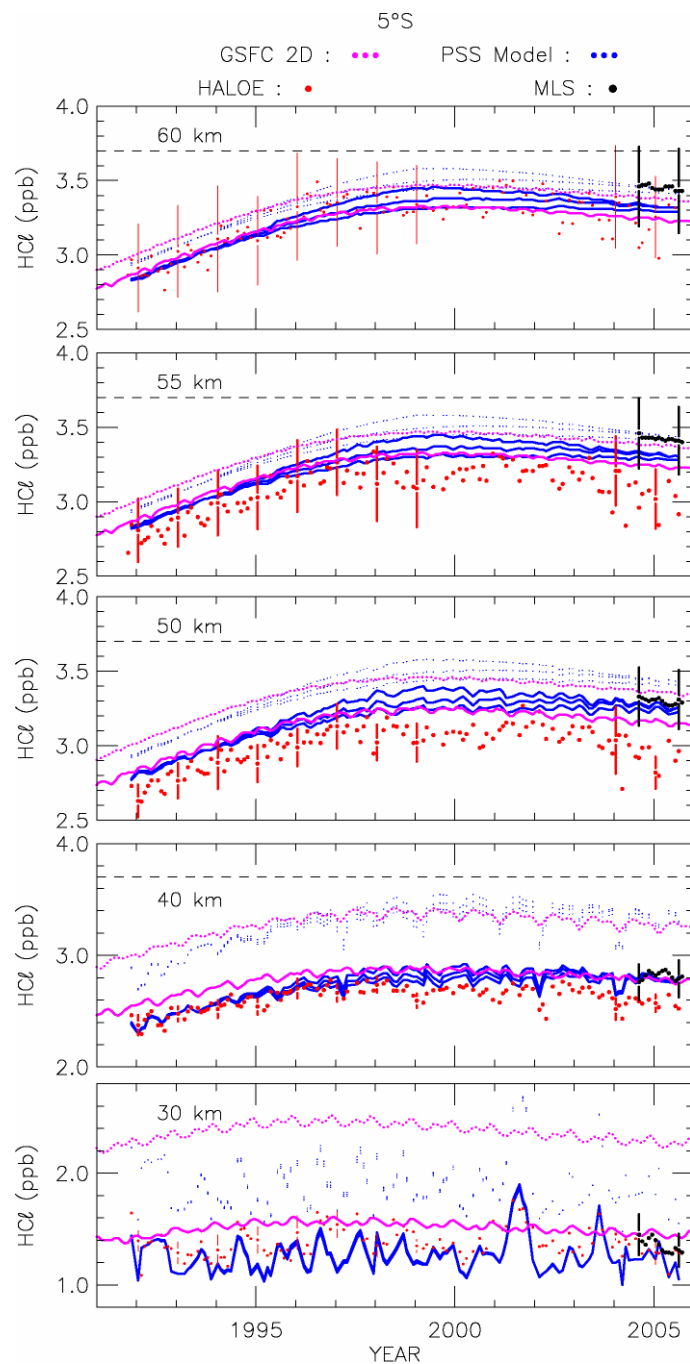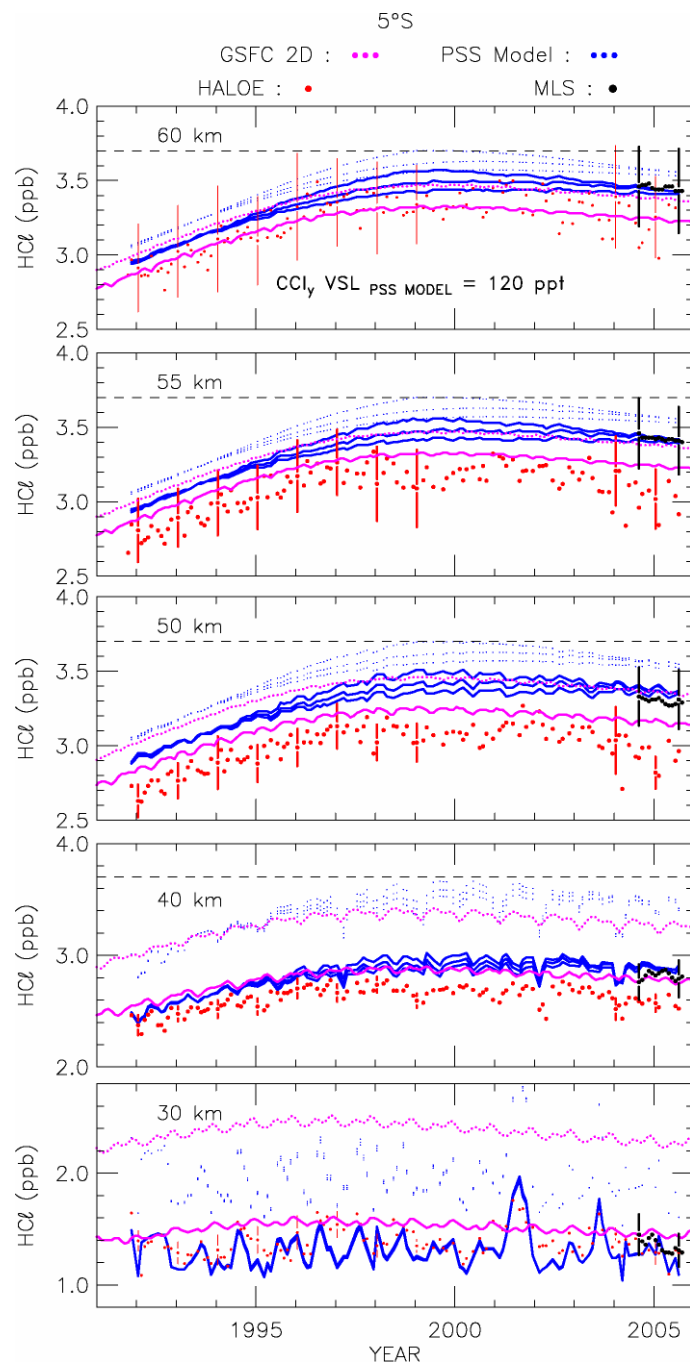
– GSFC model: WMO Cl_y
– PSS model: WMO Cl_y + 120 ppt

35°S

GSFC 2D : PSS Model :
HALOE : MLS :

35ºS

- **GSFC model: WMO Cl$_y$**
- **PSS model: WMO Cl$_y$**

35ºS

GSFC 2D : •••   PSS Model : •••

HALOE : •   MLS : •

60 km

CCl_y VSL $_{PSS\ MODEL}$ = 120 ppt

55 km

50 km

40 km

30 km

YEAR

35ºS
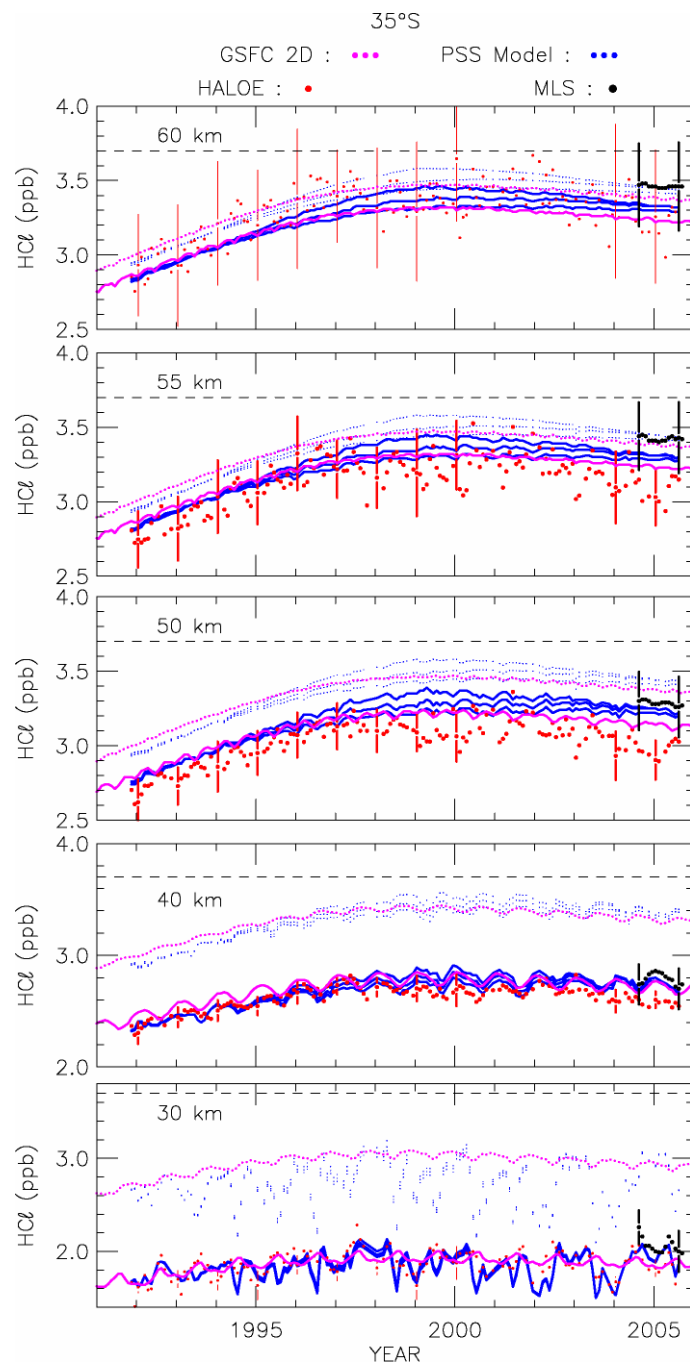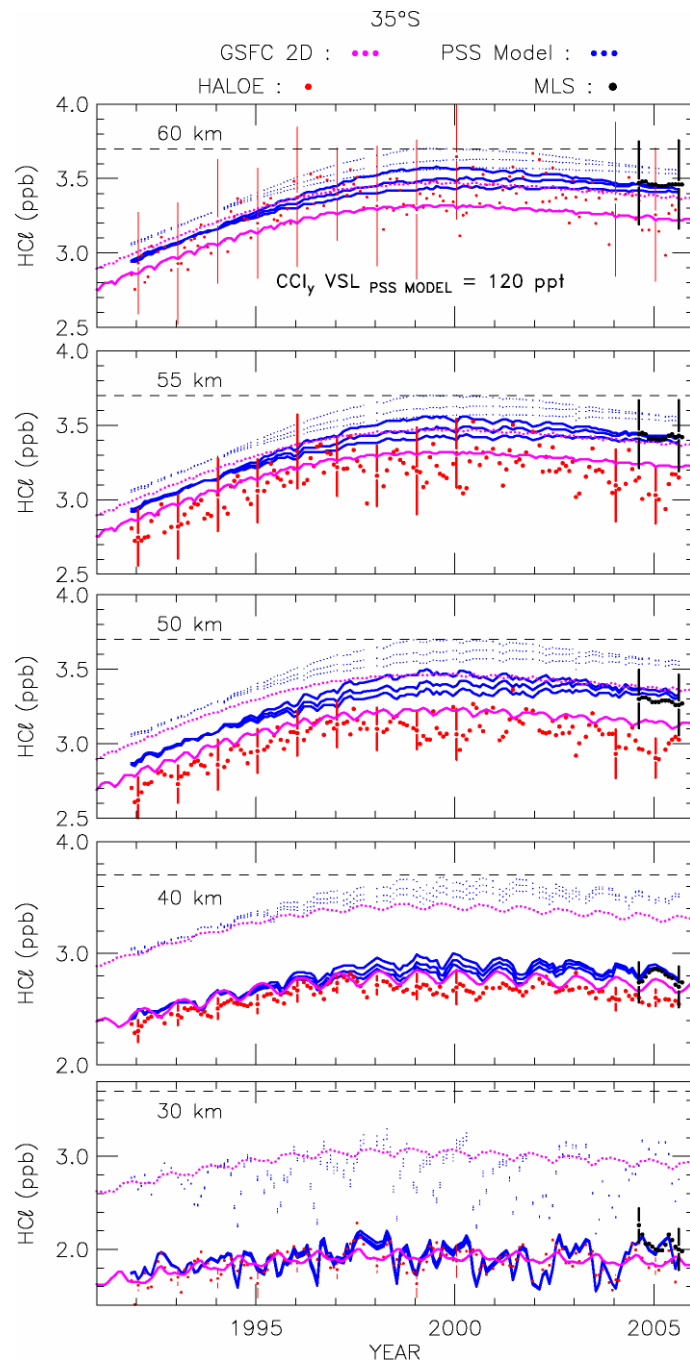
- **GSFC model: WMO Cl_y**

- **PSS model: WMO Cl_y + 120 ppt**
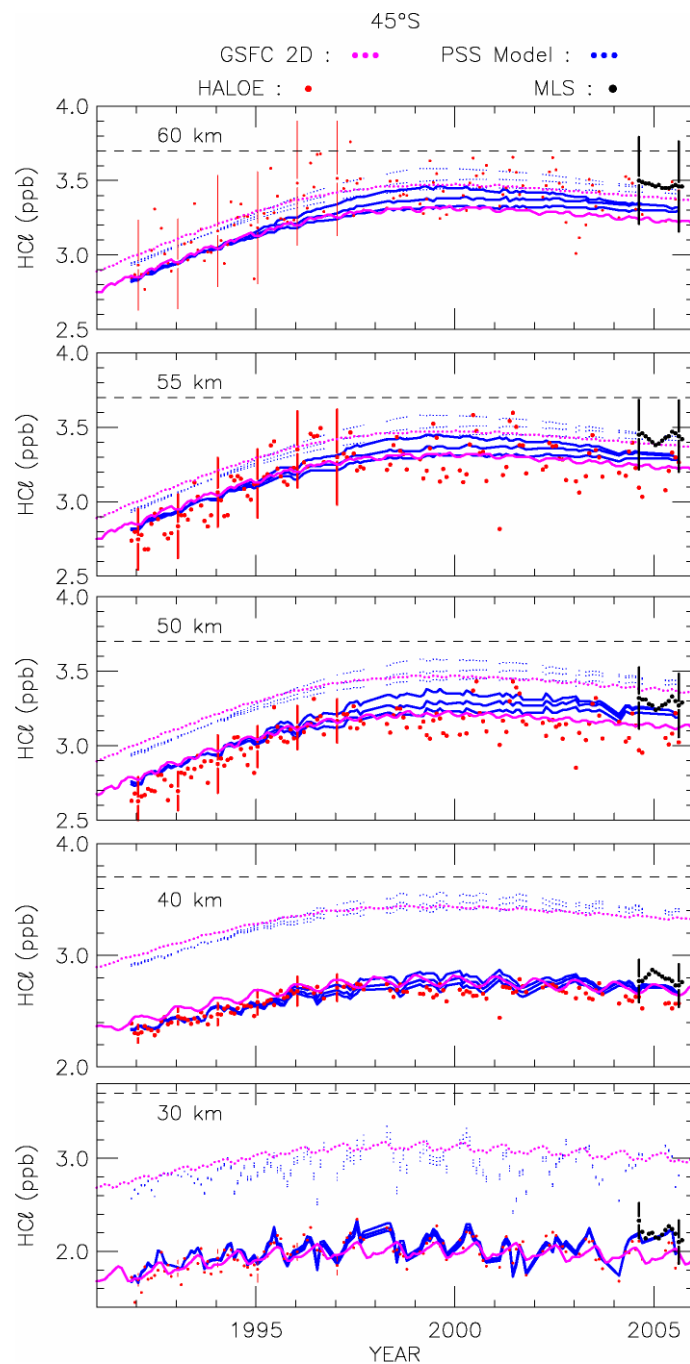
45°S

- **GSFC model: WMO Cl$_y$**
- **PSS model: WMO Cl$_y$**

45ºS
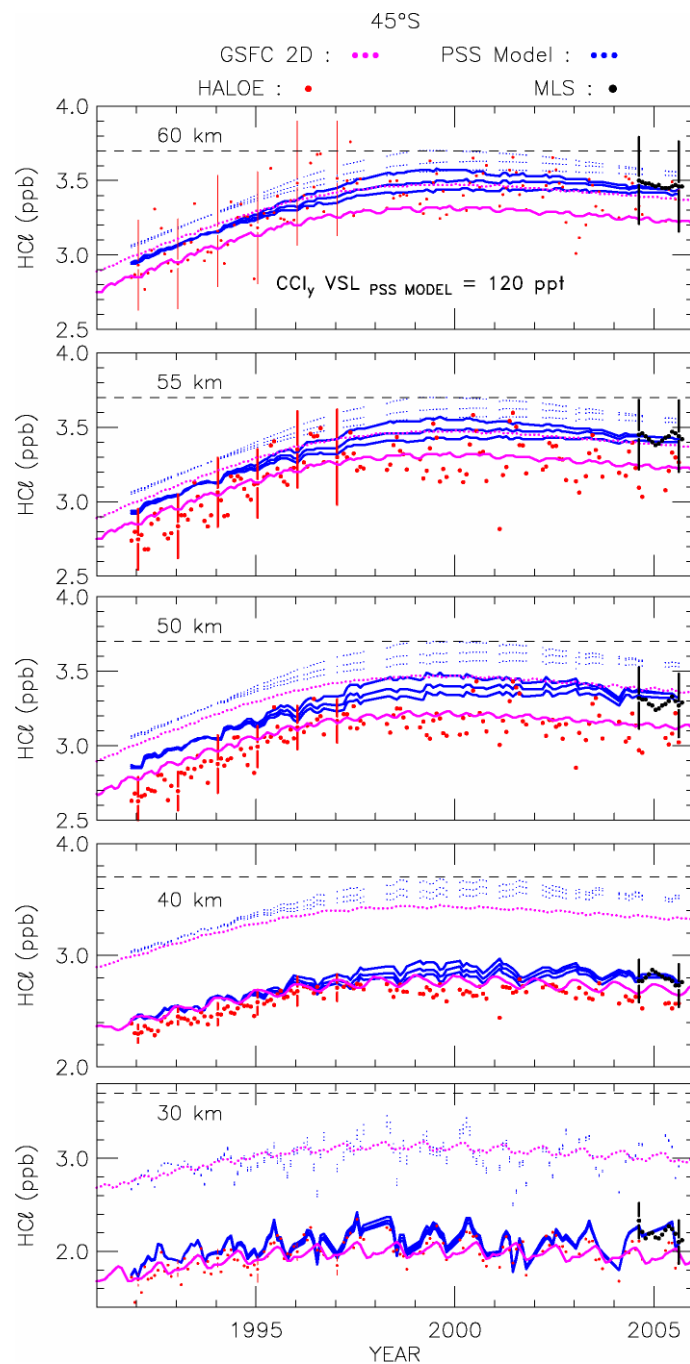
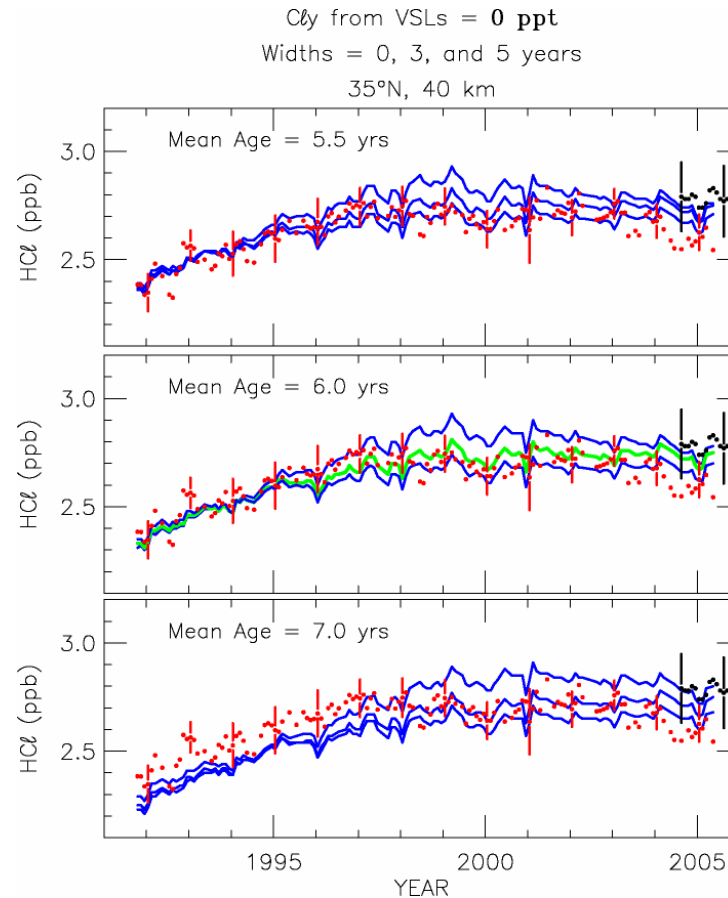– **GSFC model: WMO Cl$_y$**

– **PSS model: WMO Cl$_y$ + 120 ppt**

Sensitivity of HCl to mean age and width

35ºN, 40 km, No VSL Contribution

Darryn Waugh: Mean age likely 6 (+1, −0.5) yrs
          H_2O tape recorder & HCF134 data indicate width of 2 to 3 yrs

John Anderson: HALOE HF consistent with age = 6 yrs & width = 3 yrs ⇒ **GREEN CURVE**

Sensitivity of HCl to mean age and width

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Sensitivity of HCl to mean age and width

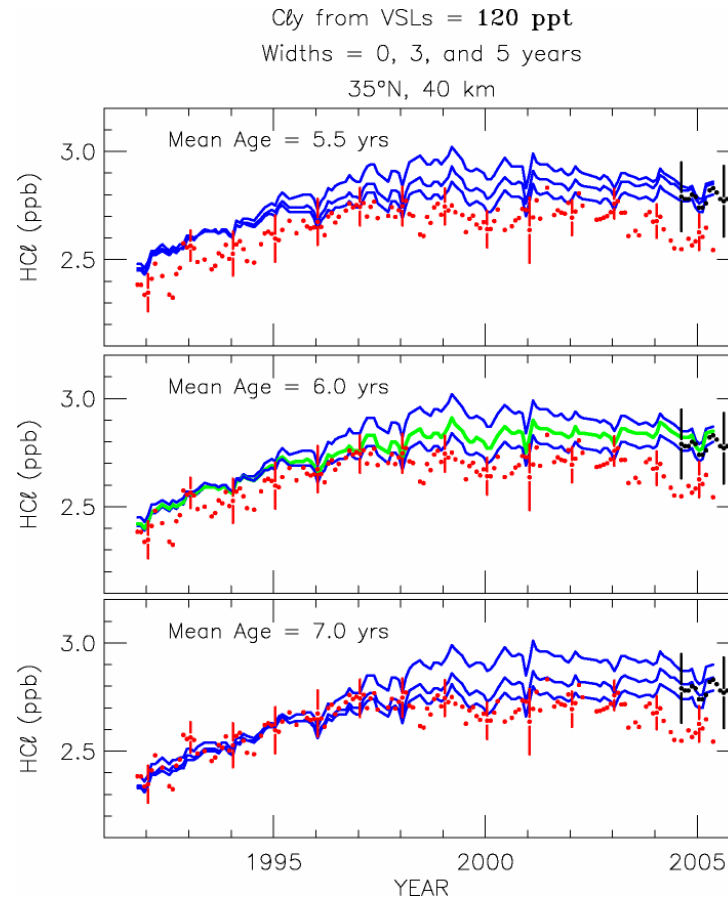## 35⁰N, 55 km, No VSL Contribution

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Sensitivity of HCl to mean age and width

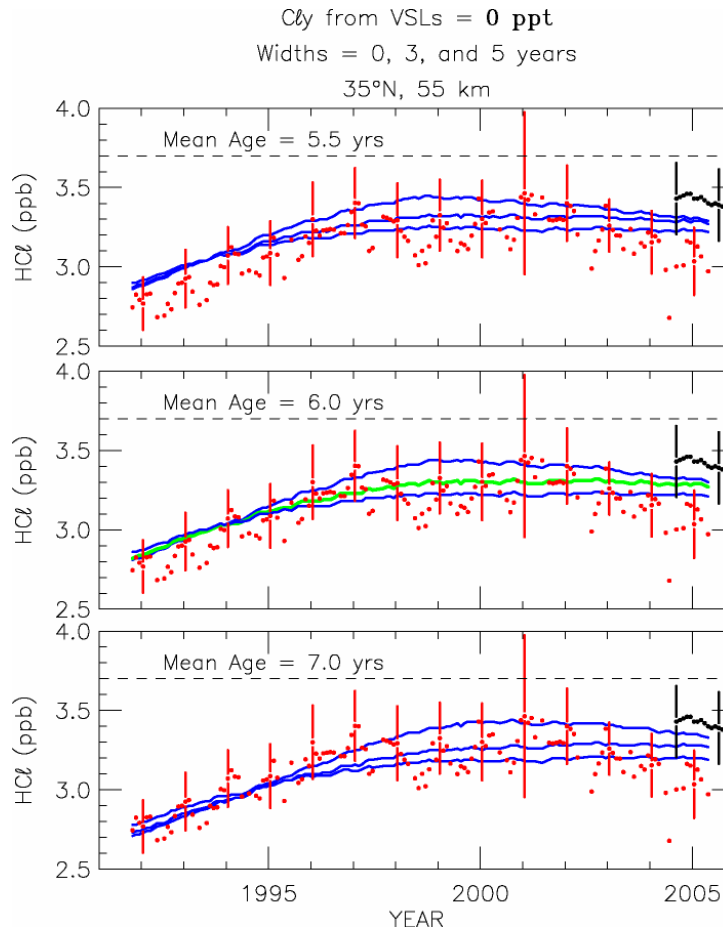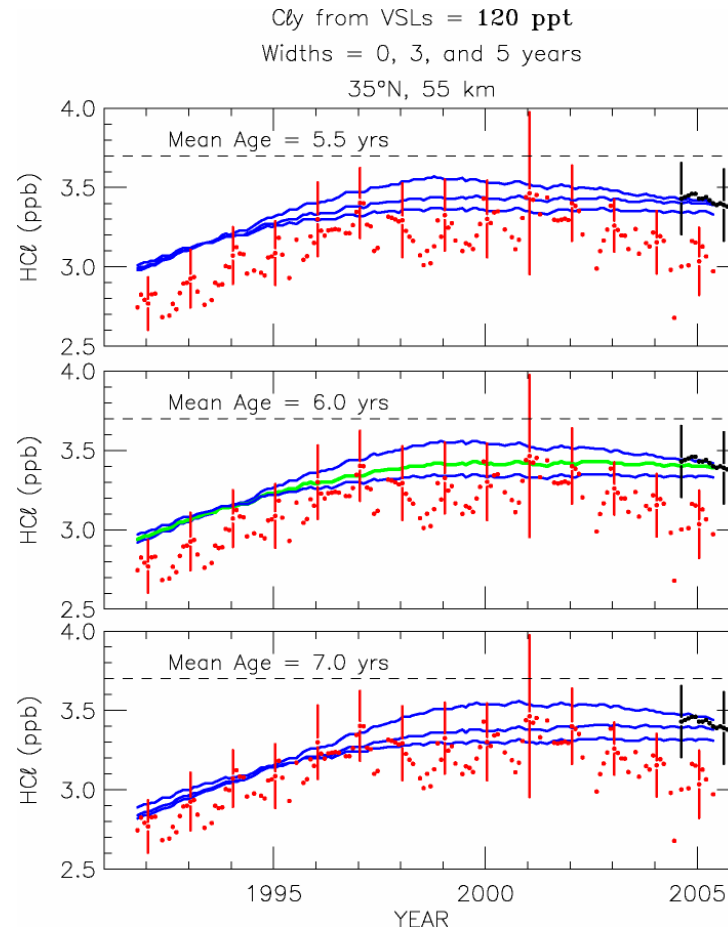## 35⁰N, 55 km, 120 ppt VSL Contribution

Darryn Waugh: Mean age likely 6 (+1, −0.5) yrs
H_2O tape recorder & HCF134 data indicate width of 2 to 3 yrs

John Anderson: HALOE HF consistent with age = 6 yrs & width = 3 yrs ⇒ **GREEN CURVE**

- Model HCl in middle to upper stratosphere depends on:
  - assumed Cl_y time series
  - mean age of air
  - <u>width</u> of the "age of air spectrum", particularly between ~1997 and 2002
- Assuming a mean age of ~6 years and a width of 2 to 3 years:
  - UARS **HALOE HCl** generally consistent with **WMO Cl_y loading**
  - Aura **MLS HCl** generally consistent with **~120 ppt contribution from VSL species to Cl_y**
- VSL contribution to Cl_y is an area of active research :
  - Chapter 2, Ko, Poulet et al., "Orange Book" (WMO/UNEP 2002 Ozone Assessment)
  - Quantification may require accurate measurements of HCl and VSL compounds in UT/LS
  - HCl solubility suggests Cl_y from VSL compounds might be "washed out"
  - Heterogeneous liberation of HCl from aerosols could "interfere with aerosol wash out of HCl"
  - Candidate VSL chlorine compounds:

Species	τ (days)	Cl content (ppt)
$C_2H_4Cl_2$	70	~ 30
$CHCl_3$	150	~ 30
CH_2Cl_2	140	~ 26
C_2Cl_4	70	~ 12

  - These species, or their decomposition products, *could* deposit Cl_y just above the tropopause

ODP (Ozone Depletion Potential) of bromine is quite sensitive
to the Cl_y abundance in the lowermost stratosphere

Extra Material To Follow

HCl Validation

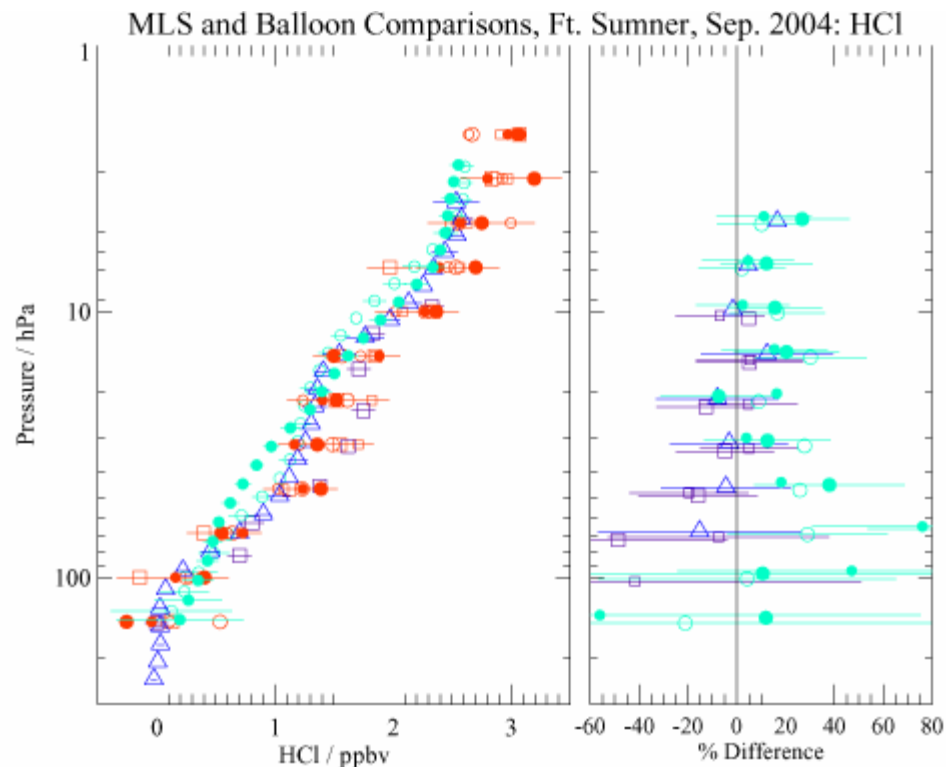MLS and Balloon Comparisons, Ft. Sumner, Sep. 2004: HCl

Fig. 17. Similar to Figure 8, but for HCl data. Left Panel: this compares MLS (red symbols) to MkIV (blue triangles) and FIRS-2 (cyan) profiles on Sep. 23/24 2004. Also shown (purple squares) is the ALIAS–II Sep. 17 (*in situ*) HCl profile retrieval, to be compared to the MLS values (red squares) for that day. Right Panel: Percent differences (for MLS minus balloon data) are shown, with symbols referring to the balloon measurements mentioned in the left panel caption. Error bars give twice the random error in these differences.
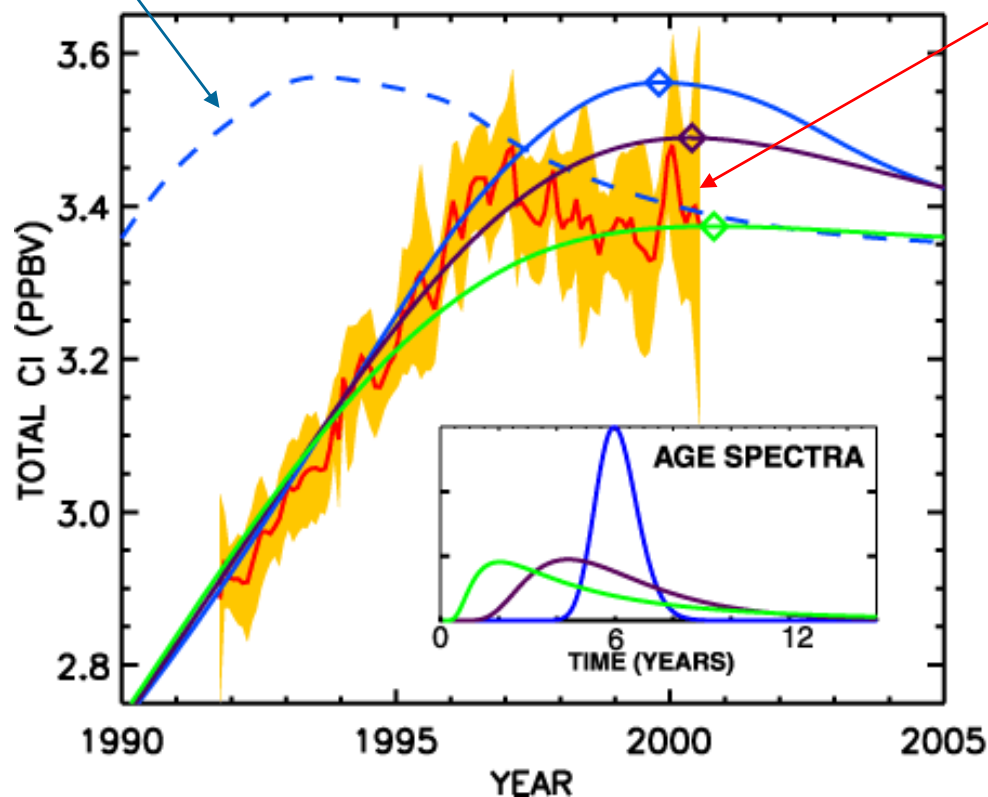
HCl Validation

MLS vs. FIRS2 HCl for 9/20/05, 36.15°N 250.55°W
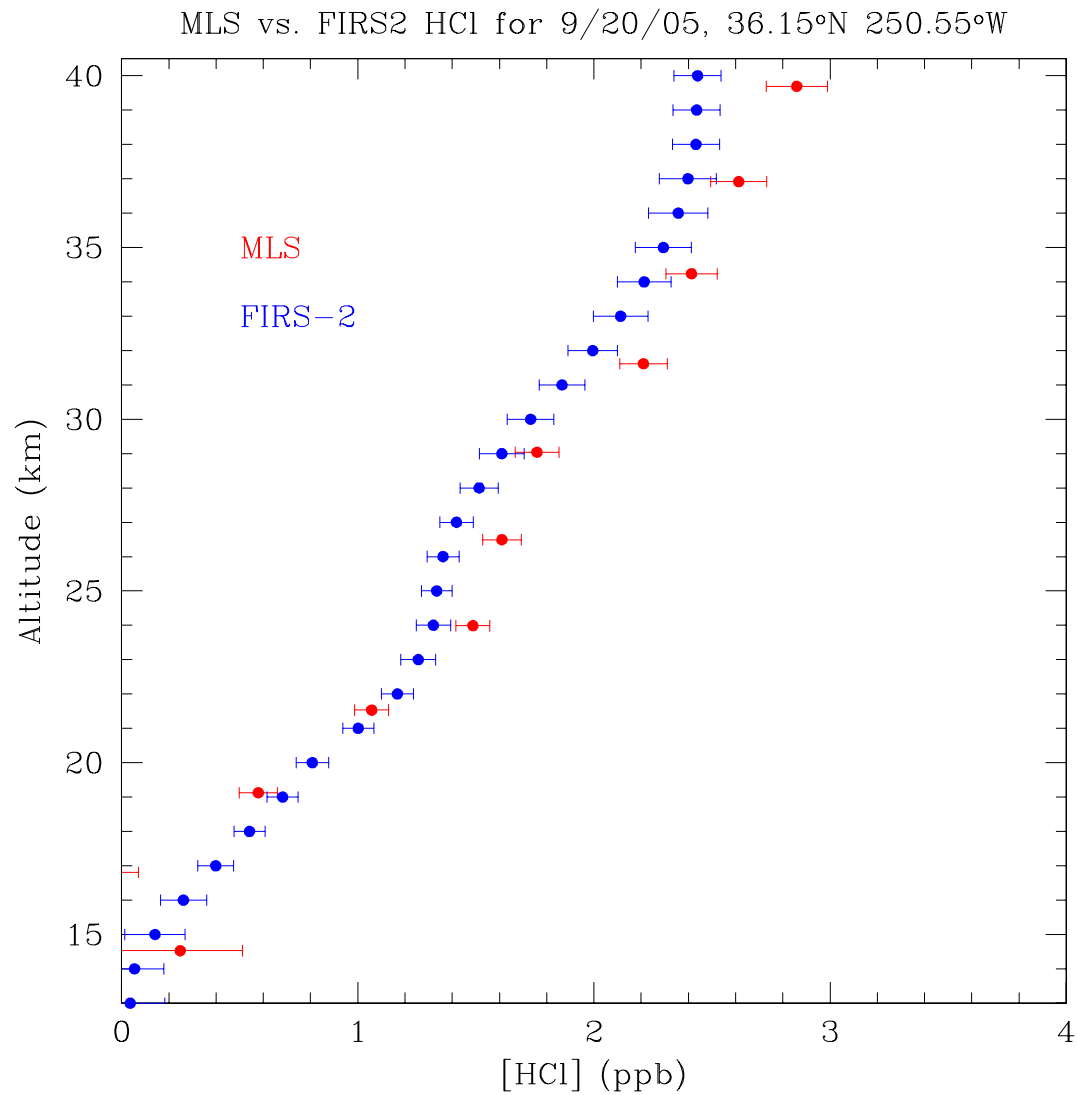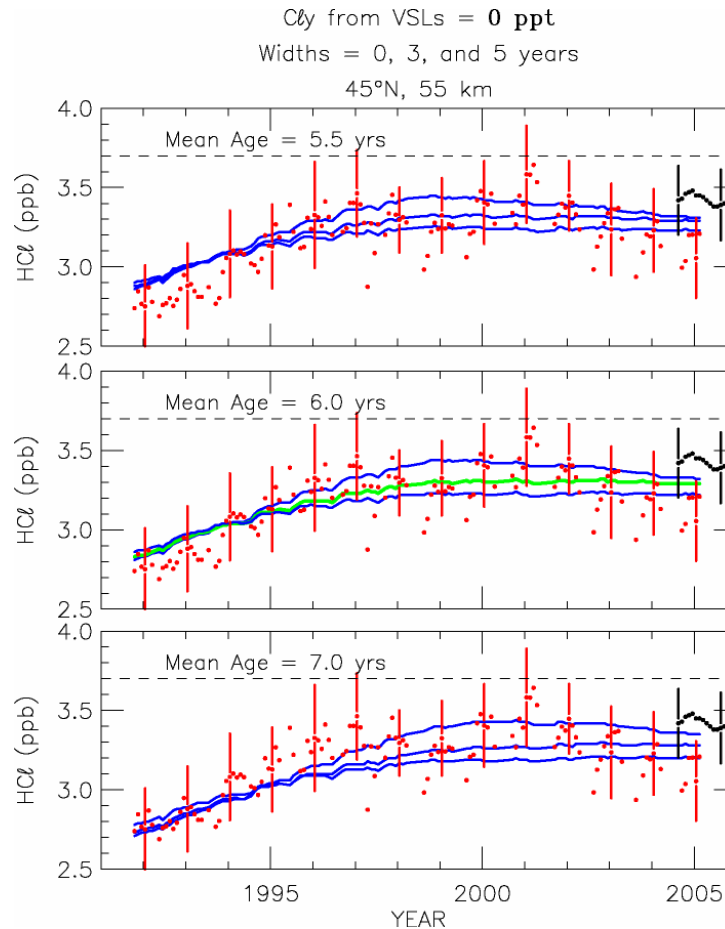
Waugh et al., GRL, 2001

Comparison of CCl$_y$ inferred from HALOE HCl, 0.46 mbar, with expected time series for CCl$_y$ with mean age of **6 years**, but various age spectra, as indicated.

Note: CCl$_y$ peaks just below 3.6 ppb in this analysis

Sensitivity of HCl to mean age and width

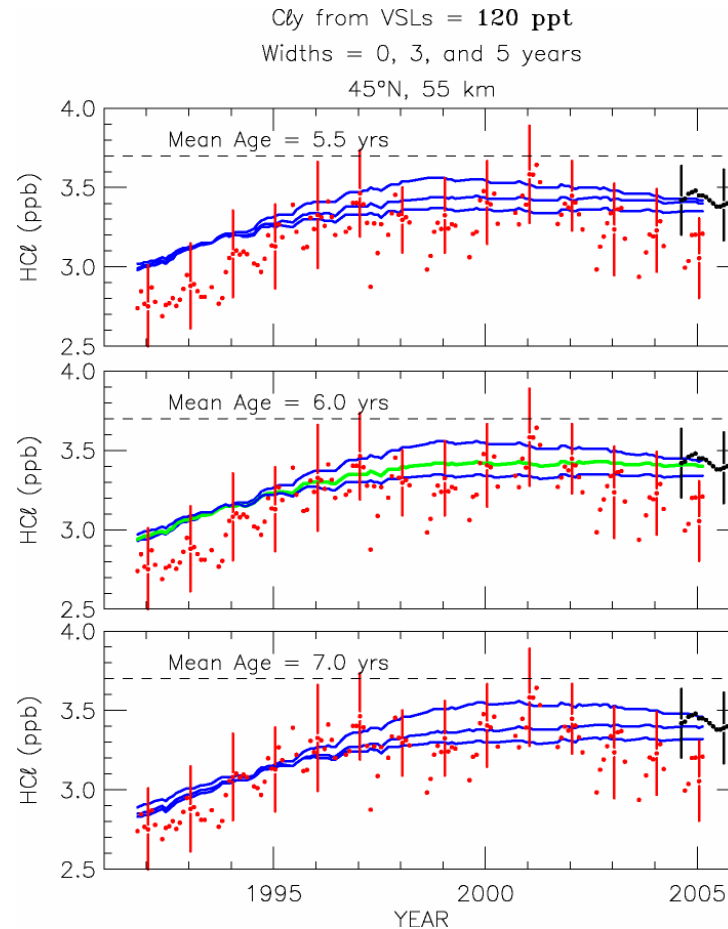## 45ºN, 55 km, No VSL Contribution

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Sensitivity of HCl to mean age and width

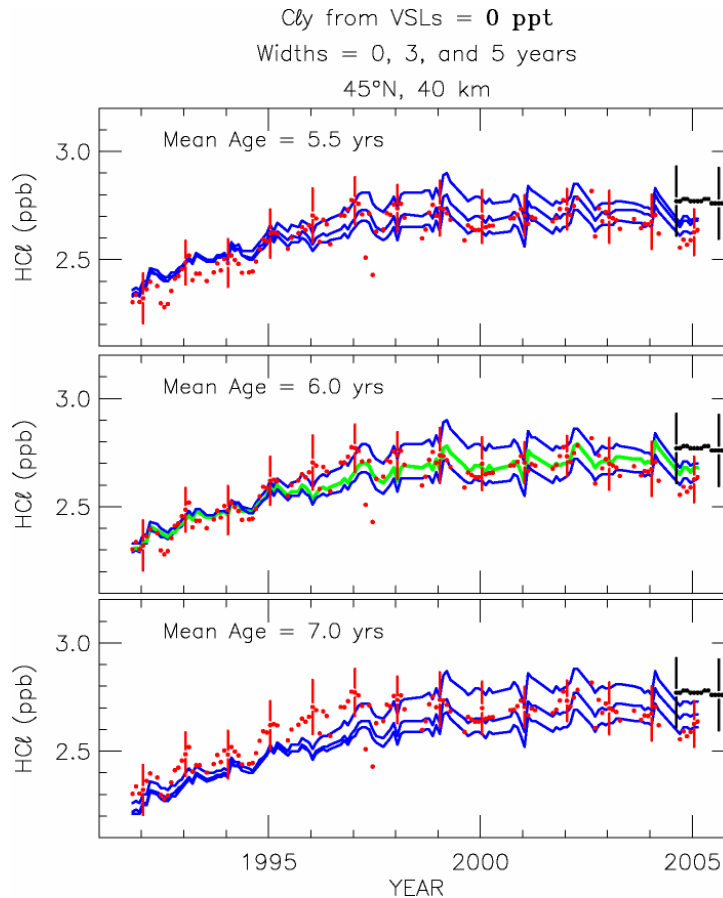## 45ºN, 55 km, 120 ppt VSL Contribution

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John Anderson: HALOE HF consistent with age = 6 yrs and width = 3 yrs

Sensitivity of HCl to mean age and width

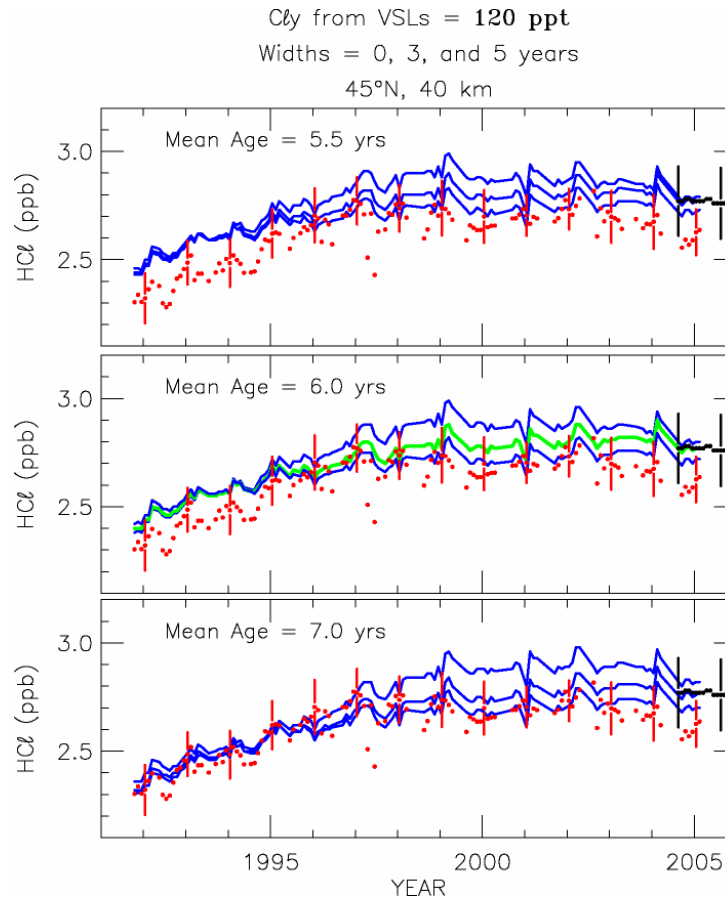## 45ºN, 40 km, No VSL Contribution

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H_2O tape recorder & HCF134 data indicate width of 2 to 3 yrs

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Sensitivity of HCl to mean age and width

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